DEGRADATION OF DISLODGEABLE AZINPHOS-METHYL (GUTHION) RESIDUE ON PEACH FOLIAGE AFTER LOW VOLUME APPLICATION IN STANISLAUS COUNTY, CALIFORNIA, 1983

by

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SUMMARY

Foliage from five peach orchards of two growers in Stanislaus County, California, was sampled before, immediately after, at 24, 48, and 72 hours, and at 7, 14, 21, and 31 days after application of azinphos-methyl (Guthion). Two orchards were treated with Guthion in June with 1 pound active ingredient (a.i.) per acre in 100 gallons of water. The remaining three orchards were treated in July with 1.5 pounds a.i. per acre in 100 gallons of water. (This was not the first application of Guthion to these three orchards in this growing season.) Sampling and subsequent analysis was conducted to evaluate the potential exposure of field workers to hazardous residues. Leaf discs were collected from four sides of the trees sampled and dislodgeable residues of Guthion and Guthion oxon analogue (GOA) were determined by gas-liquid chromatography. The highest levels of Guthion seen as averages for duplicate samples were 1.56 ug/cm^2 (60 hours) in an orchard treated with 1 pound a.i./acre and 3.14 ug/cm^2 (7 days) for an orchard treated with 1.5 pounds a.i./acre. No GOA was detected during the course of this study at the 1 pound a.i. rate. At 1.5 pounds a.i./acre, a maximum averaged level of 0.042 ug/cm^2 (14 days) was detected. Mean residue levels of Guthion plus GOA did not exceed a calculated safe level of 1.6 ug/cm^2 at the 1 pound a.i. rate. For the 1.5 pounds a.i. rate, mean residue levels exceeded the 1.6 ug/cm^2 level even after 31 days. There were considerably more days in the July study in which the air temperature was 95°F and above than in the June study. Total oxidant levels were also higher in July than in June. All these factors (application rate, dilution rate, weather conditions, and oxidant levels) may affect the residue levels. Further study is needed.

INTRODUCTION

Organophosphate pesticide residues and their corresponding oxidation products are known to cause field worker illnesses through reduction of cholinesterase enzyme activity when toxic exposure occurs due to early reentry into treated fields, orchards, or groves. The severity of such toxic exposure, and any possible resulting illness, is dependent upon the extent of prolonged or repeated contact with dislodgeable foliar residue, and to a presumably lesser extent, contact with contaminated particles of the soil (1, 2). Also of prime importance, is the quantity and toxicity of residues present at the time of exposure. It is often true that oxidation products are more toxic than the pesticide from which they are derived. In the case of Guthion oxon analogue (azinphos-methyl oxon, GOA), the highly toxic oxidation product of Guthion (azinphos-methyl), the ratio of toxicity between the two could be as high as 60 to 1 (3). Guthion alone has an oral LD_{50} in the rat of 11 to 16.4 mg/kg and a dermal LD_{50} in the rat of 220 mg/kg (4). Guthion is a Toxicity Category I pesticide based on oral LD_{50} and is a California Restricted Material.

To reduce the potential for field worker illnesses due to early reentry into treated areas where toxic levels of pesticide residue are present, the California Department of Food and Agriculture (CDFA) has established reentry intervals for selected pesticides which restrict entry into an orchard or field prior to the time when residues reach a level calculated to be safe. Most current reentry intervals were empirically set in 1971, and were both application rate and commodity dependent (5). Reentry intervals for additional pesticides have been added since that time.

In order to provide greater protection to field workers and, where it can be done safely, greater flexibility to growers, the Department has now begun an evaluation of the existing reentry intervals and might make changes in the present regulations based on the results of newer reentry studies. Factors now being considered include the effects of differing application methods and equipment, the effects of humidity and irrigation practices on pesticide degradation, and the effects of high temperatures and varying levels of airborne oxidants on the degradation of pesticide residues and the production of their oxon analogues. This study was undertaken in the early summer of 1983 by the Northern Field Team of the Worker Health and Safety Unit to provide data to be used in the evaluation of the current 14-day reentry interval for Guthion on peaches and nectarines.

The application of Guthion to peaches in Stanislaus County occurs primarily in the spring and early summer for the control of the Oriental Fruit Moth, the Peach Twig Borer, and various aphids, thrips, and scale insects. The formulation used at present is a 50 percent wettable powder which is tank-mixed in water and is normally applied by ground application equipment. The tank mix may or may not include adjuvants or other pesticides.

Low volume pesticide application methods have been in use in California citrus growing operations since at least the early 1970's (6). More recently, this practice has been followed for other tree fruits and for

pesticides such as ethyl parathion and Guthion (7-9). Current application technology allows the grower to use far less water in a tank mix than would be used for a more dilute, full coverage spray application, therefore allowing a significant savings of time, a reduction in equipment wear, and a lowering of total pest management costs. In addition, low volume applications have been shown to result in higher pesticide residues on fruit and foliage than applications of an equal amount of active ingredient per acre in a more dilute spray mixture; total coverage and uniform distribution, however, are less likely to be achieved (6). These higher residue levels resulting from low volume applications could be expected to potentially increase field worker health risks during reentry into treated orchards or groves to perform work activities that result in substantial contact with foliage, fruit, or soil dust.

The low volume application of Guthion on peaches is allowed by a clause on the current label which states, in effect, that the maximum rate of 6 pounds of a 50 percent Guthion formulation which can be applied in a full coverage spray containing up to 800 gallons of water per acre, can also be applied in a more concentrated spray. No lower limit of water to be used for mixing is recommended on the label.

METHODS

A list of peach growers in Stanislaus County was provided by personnel from the Stanislaus County Department of Agriculture. Growers were then approached on an individual basis by State personnel to request cooperation for this study. Cooperation of a grower (Grower One) near Salida, north of Modesto, was obtained in early June and sampling of treated orchards began soon afterward. A second grower (Grower Two) was located near Hughson, south of Modesto, and sampling was initiated in early July in conjunction with his final Guthion application prior to harvest.

Two orchards owned by Grower One were selected for study. The older of the two was flood irrigated, while the newer was irrigated by sprinkler. Both were irrigated once during the sampling period. Guthion was applied to the sprinkler irrigated orchard shortly after noon on June 6. The flood irrigated orchard was treated between 3:00 and 4:00 a.m. on June 7. The application rate for both orchards was 2 pounds of 50 percent Guthion per 100 gallons of water per acre or 1 pound a.i. per acre.

One month later, three orchards belonging to Grower Two plus an untreated control orchard were sampled. All were flood irrigated, and each was irrigated once during the sampling period. The only major difference between the treated fields was in variety. The varieties sampled were: McKnights, Monacos, and Carolines. The application rate used by Grower Two was 3 pounds of 50 percent Guthion per 100 gallons of water per acre or 1.5 pounds a.i. per acre. All three orchards received the same application rate with treatment occurring in the early morning of July 8. In addition, each of the treated orchards was cultivated once during the sampling period.

Foliar residue samples were collected using methods as described by Gunther, et al., (10) and Iwata, et al., (1). Sampling intervals were set

for the following times: Immediately post-application, 24 hours after application, 48 hours, 72 hours, 7 days, 14 days, 21 days, and the final samples were collected 31 days post-application. Pre-application samples were collected to determine levels of Guthion or other pesticides that were present prior to application. This schedule was followed precisely for the samples of Grower Two, but was altered slightly for the orchards of Grower One. For that application, samples from one orchard were staggered by 12 hours in order that samples for each treatment could be collected at the same time. After collection, samples were sealed in glass jars with foil-lined lids and kept on ice until delivery to the laboratory.

Sampling rows for each orchard were determined and marked prior to application when pre-application samples were obtained. Each successive sampling was taken from the same trees. One 2.54 cm leaf disc was punched from each "side" of a sample tree on both a perpendicular and a parallel axis to the row, for a total of four punches per tree. Each disc was punched at a height of five to six feet above ground level to minimize interference from irrigation. Fifteen trees were punched for a total of 60 discs per sample, and two rows or samples were obtained from each orchard at each sampling interval.

Foliar dislodgeable residues were analyzed using standard method number 27.0 as established by CDFA's Chemistry Laboratory in Sacramento. Residues were extracted from the leaf disc surfaces in a water/Surten solution. Accumulated water was then extracted with dichloromethane and evaporated. Known volume solutions were made with ethyl acetate and analyzed by gas-liquid chromatography. Specific equipment conditions for Guthion and GOA were as follows:

Column	4% OV-1 on 100/120 gaschrome Q in 3 ft. x 2mm glass column
Oven Temperature	220 degrees
Detector	NPD
Carrier Gas	Nitrogen at 30 ml/min.
Chart Speed	.5 cm/min.
Range	1 x 1
Retention Time	8.74 min.
Injector Temperature	230 degrees

RESULTS

Pertinent characteristics of each of the applications for Growers One and Two are shown in Table I, including brand name, application rate, and quantity of active ingredient per acre. Climatological data including temperature and levels of airborne oxidants (nitrous oxides, ozone, and total [$NOx+0_3$]) are presented in Tables II and III for Growers One and Two, respectively. Table IV presents mean values of Guthion and GOA residues for the duplicate samples taken in each orchard.

Figure 1 presents mean value degradation rates for the flood and sprinkler irrigated orchards of Grower One. Figure 2 shows degradation rates for

the Monaco and McKnight varieties of peaches for Grower Two. Figure 3 presents degradation in the Caroline variety and the control orchard. All three figures show the mean daily temperature and total oxidant level for the respective sampling periods. Degradation rates can be compared with the 1.6 ug/cm^2 calculated safe level for Guthion plus GOA as determined by Knaak and Iwata (3) and shown on each graph.

The highest mean levels of dislodgeable Guthion residue seen for Grower One were in the 60- and 72-hour samples with 1.56 and 1.29 ug/cm^2, respectively. No GOA was detected during the sampling period. At no time during the sampling period did the averaged and combined residues exceed the 1.6 ug/cm^2 level for Guthion plus GOA. One unaveraged sample for the flood irrigated field did, however, reach 1.66 ug/cm^2 Guthion in the immediately post-application sample, but this had degraded to below 1.6 ug/cm^2 within 12 hours after application.

No obvious effect was seen, in this situation, in the degradation rate based on the type of irrigation used for the two orchards of Grower One. The flood irrigated orchard had been irrigated between the 60-hour and the 7-day sample and the sprinkler irrigated orchard was irrigated between the eighth and fifteenth day. While the flooded orchard had initially higher residue levels, the residue present for the last three weeks of sampling was equal to or below that determined for the sprinkler irrigated orchard. Any true comparison between these two types of irrigation and their effect on residue degradation are outside the scope of this study.

The residue levels determined for the orchards of Grower Two were significantly higher than those of Grower One during the entire sampling period. The maximum unaveraged level of dislodgeable Guthion residue present was detected on the seven-day sample for the Caroline variety at 3.59 ug/cm^2. The maximum unaveraged level of GOA was detected on the same variety at 0.049 ug/cm^2 at 14 days. The highest mean levels were determined in the same variety at the same sampling times and were 3.14 ug/cm^2 and 0.042 ug/cm^2, respectively.

For Grower Two, Guthion residue was present in all orchards including the control prior to Guthion applications studied here. All three orchards had been treated five weeks prior to the start of this study with 1.2 pounds a.i./acre Guthion. The control orchard was treated at approximately the same time. The average level of Guthion present before application was 0.84 ug/cm^2 and for GOA, 0.011 ug/cm^2. Phosalone (Zolone) residue was also detected in the control and in the Monaco orchard prior to application at an average level of 0.009 ug/cm^2.

The three treated orchards for Grower Two were irrigated late in the sampling period, but the control orchard was irrigated within two days of the application date for the other orchards. The Monaco variety received irrigation just before the seven-day samples were collected. The Carolines and the McKnights were irrigated shortly before the final samples were collected.

The quantity of dislodgeable Guthion residue present in the three treated orchards exceeded the 1.6 ug/cm^2 calculated safe level for Guthion plus

GOA immediately after application, and did not fall below this level by the end of the 31 days of sampling. The quantity of dislodgeable GOA, however, did not exceed a calculated safe level of 0.05 ug/cm^2 for GOA alone (3) at any time.

DISCUSSION AND CONCLUSIONS

For all treated orchards, the highest levels of Guthion residue were detected at least 60 hours and as much as 14 days after the finish of application. This was also true for samples of the untreated control orchard. For the treated orchards of Grower Two, this was originally thought to be due to some type of interaction between Guthion and Phosalone or one of the fungicides used earlier in the year, but since a similar but less dramatic result was seen for the orchards of Grower One, this might not be the case. Also, only minute traces of Phosalone were present prior to treatment with Guthion. Laboratory problems would also be ruled out, since the samples for Grower One were run a month before those of Grower Two. Regardless of the source of this increase, the orchards of Grower One would appear safe to enter within 24 hours, while the orchards of Grower Two did not reach the calculated safe residue level even after 31 days. Even the control orchard, which was not treated with the others, had one averaged sample showing 1.6 ug/cm^2. Even if the level of Guthion in the presamples or in the control were subtracted from the Guthion results, there would still be residue levels exceeding 1.6 ug/cm^2 at 7 days and at 31 days, respectively, in both cases.

The most likely source of increasing levels of dislodgeable Guthion residue as time progressed can be found by examining the relationship between temperature and residue level. Examination of Figures 1 through 3 shows graphically that in conjunction with periods of temperature in the high 90's or above, the residue level on foliage was seen either to rise or remain at a uniform level and not reduce with time. The graphs also show that total oxidants present closely follow changes in temperature. Statistical evaluation shows a highly significant correlation (1%) for the close relationship between temperatures and Guthion residue. Likewise, there is a highly significant correlation (1%) between temperature and total oxidant level. No such close relationship could be demonstrated for the relationship between total oxidant and Guthion residue levels. This information supports the belief that any adjustment to reentry intervals needs, in part, to be based on ambient temperatures.

Peach growers in Stanislaus County lack a certain degree of flexibility in regard to the use of Guthion late in the growing season to control the Oriental Fruit Moth and the Peach Twig Borer. Applications must be accurately timed in conjunction with periods of moth flight to be successful. Although there is a 24-day preharvest interval, a shortened reentry period would allow field workers to complete work activities, such as thinning of fruit or propping of limbs, rather than having to postpone such activities while waiting for a reentry interval to expire. It is clear, however, based on the data in this study, that for low-volume applications of 1.5 pound a.i./acre or more and high temperatures, a reentry interval of 14 days may not be long enough.

To allow earlier reentry when conditions are safe, a graduated system of varying reentry intervals based on dilution and quantity of active ingredient per acre has been proposed. This might be the best method of providing flexibility to growers while ensuring worker safety. Under this system, the reentry interval would increase as the quantity of Guthion per acre increased, but would decrease as the quantity of water used for dilution increased. For the full coverage, very dilute sprays with low levels of active ingredient, a reentry interval of less than 14 days, might be possible. This would allow greater grower flexibility, however, efficacy would also need to be considered.

It would be useful to note that several peach growers approached concerning cooperation for this study would have provided orchards if they had not already completed their final Guthion applications of the season. Future studies should be arranged in May or early June to have a wider selection of growers using varied application rates.

It was learned during this study that field workers who enter treated orchards for "propping" may have substantial contact with treated foliage. This work activity involves placing of boards to support limbs that are heavy with fruit. The activity occurs in late June or early July so it is required during times when orchards may have been treated with Guthion. No worker exposure data on this work activity has been found.

Table I - Characteristics of Guthion Applications to Peach Orchards Sampled in Stanislaus County, 1983

Grower	Orchard	Application Date	Finish Time	Brand of Guthion	EPA Reg. Number	Application Rate/Acre	Active Ingredient Per Acre (lbs)
1	Flood	June 7, 1983	By 4:00 a.m.	Mobay Guthion 50%	3125-301 AA	2 lbs./100 gal.	1
	Sprinkler	June 6, 1983	By 2:00 p.m.	Mobay Guthion 50%	3125-301 AA	2 lbs./100 gal.	1
2	Monacos	July 8, 1983	By 6:00 a.m.	Mobay Guthion 50%	3125-301 AA	3 lbs./100 gal.	1.5
	Carolines	July 8, 1983	By 9:30 a.m.	Mobay Guthion 50%	3125-301 AA	3 lbs./100 gal.	1.5
	McKnights	July, 8, 1983	By 9:30 a.m.	Mobay Guthion 50%	3125-301 AA	3 lbs./100 gal.	1.5

Table II - Climatological Data Collected For the Sampling Period for Grower One

Month	Date	Temperature (°F)[1] Max.	Min.	Mean	Rainfall	Oxidant Level (PPHM)[2] NO_x	O_3	Total[3]
June	7	94	71	83	No rainfall	2.6	3.3	5.9
	8	87	58	73	was recorded	2.5	2.0	4.5
	9	89	57	73	during sam-	2.8	1.6	4.4
	10	88	56	72	pling period.	2.8	0.6	3.4
	11	80	54	67		2.7	2.3	5.0
	12	88	54	71		3.0	1.9	4.9
	13	95	57	76		4.2	2.7	6.9
	14	93	61	77		4.0	2.1	6.1
	15	90	58	74		2.8	2.0	4.8
	16	100	61	81		2.9	2.6	5.5
	17	87	61	74		2.0	1.2	3.2
	18	84	54	69		2.6	2.0	4.6
	19	88	56	72		2.3	1.9	4.2
	20	88	57	72		2.2	1.7	3.9
	21	90	57	74		1.8	2.3	4.1
	22	93	59	76		2.1	2.6	4.7
	23	87	57	72		1.5	1.1	2.6
	24	89	55	72		2.5	1.2	3.7
	25	94	56	75		3.2	3.6	6.8
	26	94	63	79		2.3	0.8	3.1
	27	82	55	69		2.6	0.9	3.5
	28	85	54	70		2.5	1.1	3.6
	29	85	56	71		2.4	1.1	3.5
	30	83	55	69		3.2	1.1	4.3
July	1	78	60	69		2.6	0.1	2.7
	2	82	59	71		2.7	1.3	4.0
	3	91	62	77		2.7	2.3	5.0
	4	100	62	81		2.0	2.0	4.0
	5	100	68	84		2.7	2.0	4.7
	6	82	59	71		2.5	1.3	3.8
	7	77	54	66		2.5	0.9	3.4
	Means	89	58	74		2.6	1.7	4.3

1/ Weather data from the National Weather Service for Modesto.

2/ Oxidant data from the California Air Resources Board, Airmetric Division for Modesto Station.

3/ Total Oxidant = $NO_x + O_3$

Table III - Climatological Data Collected For the Sampling Period for Grower Two

Month	Date	Temperature (°F)[1/] Max.	Min.	Mean	Rainfall	Oxidant Level (PPHM)[2/] NO_x	O_3	Total[3/]
July	7	77	54	66	No rainfall	2.5	0.9	3.4
	8	80	51	67	was recorded	2.7	1.0	3.7
	9	85	55	70	during sam-	2.6	1.3	3.9
	10	94	55	75	pling period.	2.6	2.3	4.9
	11	100	62	81		3.4	2.9	6.3
	12	104	65	85		5.0	3.9	8.9
	13	107	65	86		3.9	4.3	8.2
	14	97	70	84		2.0	1.9	3.9
	15	93	66	78		2.5	2.2	4.7
	16	90	59	75		1.8	2.1	3.9
	17	82	53	68		2.0	1.3	3.3
	18	82	54	68		2.5	0.6	3.1
	19	82	55	69		2.4	0.9	3.4
	20	88	57	73		2.8	2.1	4.9
	21	94	58	76		3.8	3.5	7.3
	22	88	57	73		2.0	1.9	3.9
	23	87	57	72		2.1	3.0	5.1
	24	85	57	71		2.0	2.3	4.3
	25	84	55	70		2.4	1.3	3.7
	26	89	57	73		2.1	2.2	4.3
	27	93	60	77		1.8	2.4	4.2
	28	93	60	77		1.6	1.5	3.1
	29	94	61	78		1.9	2.4	4.3
	30	96	63	80		1.4	2.7	4.1
	31	95	62	79		1.0	2.9	3.9
August	1	98	64	81		2.4	3.2	5.6
	2	97	65	81		1.8	1.9	3.7
	3	96	65	81		1.7	1.9	3.6
	4	90	60	75		1.9	1.0	2.9
	5	93	61	77		2.8	2.4	5.2
	6	102	63	83		3.4	4.3	7.7
	7	107	71	89		2.2	4.6	6.8
	8	101	73	87		1.8	1.9	3.7
	Means	92	60	77		2.4	2.3	4.7

1/ Weather data from the National Weather Service for Modesto.

2/ Oxidant data from the California Air Resources Board, Airmetric Division for Modesto Station.

3/ Total Oxidant = $NO_x + O_3$

Table IV - Mean Values of Guthion and Guthion Oxon Analogue for Duplicate Samples Collected for Five Peach Orchards Plus a Control in Stanislaus County, 1983

		Postsample		24 Hours (12 Hours)		48 Hours (36 Hours)		72 Hours (60 Hours)		7 Days (8 Days)		14 Days (15 Days)		21 Days (22 Days)		31 Days (30 Days)	
		G[1]	GOA[2]	G	GOA	G	GOA	G	GOA	G	GOA	G	GOA	G	GOA	G	GOA
Grower 1	Flood	1.39	ND[3]	(1.45)	(ND)	(1.46)	(ND)	(1.56)	(ND)	0.88	ND	0.88	ND	0.78	ND	(0.39)	(ND)
	Sprinkler	1.24	ND	1.27	ND	1.27	ND	1.29	ND	(0.95)	(ND)	(0.88)	(ND)	(0.77)	(ND)	0.62	ND
Grower 2	Monacos	2.10	ND	2.61	ND	1.99	ND	2.64	0.020	2.48	ND	2.00	0.021	1.54	0.027	2.25	0.025
	Carolines	2.10	ND	2.38	ND	2.35	0.023	2.18	0.023	3.14	0.035	2.43	0.042	2.24	0.030	2.53	0.041
	McKnights	1.14	ND	1.65	ND	1.82	ND	1.44	ND	2.32	ND	1.44	0.029	1.72	0.022	1.86	0.022
	Untreated Control	0.56	ND	0.89	ND	0.69	ND	0.77	ND	1.64	0.017	0.86	0.018	0.76	0.018	0.82	0.028

[1] Guthion in ug/cm^2; Minimum Detectable Level = 0.002 ug/cm^2

[2] Guthion oxon analogue in ug/cm^2; Minimum Detectable Level = 0.01 ug/cm^2

[3] ND = None detected at or above the MDL

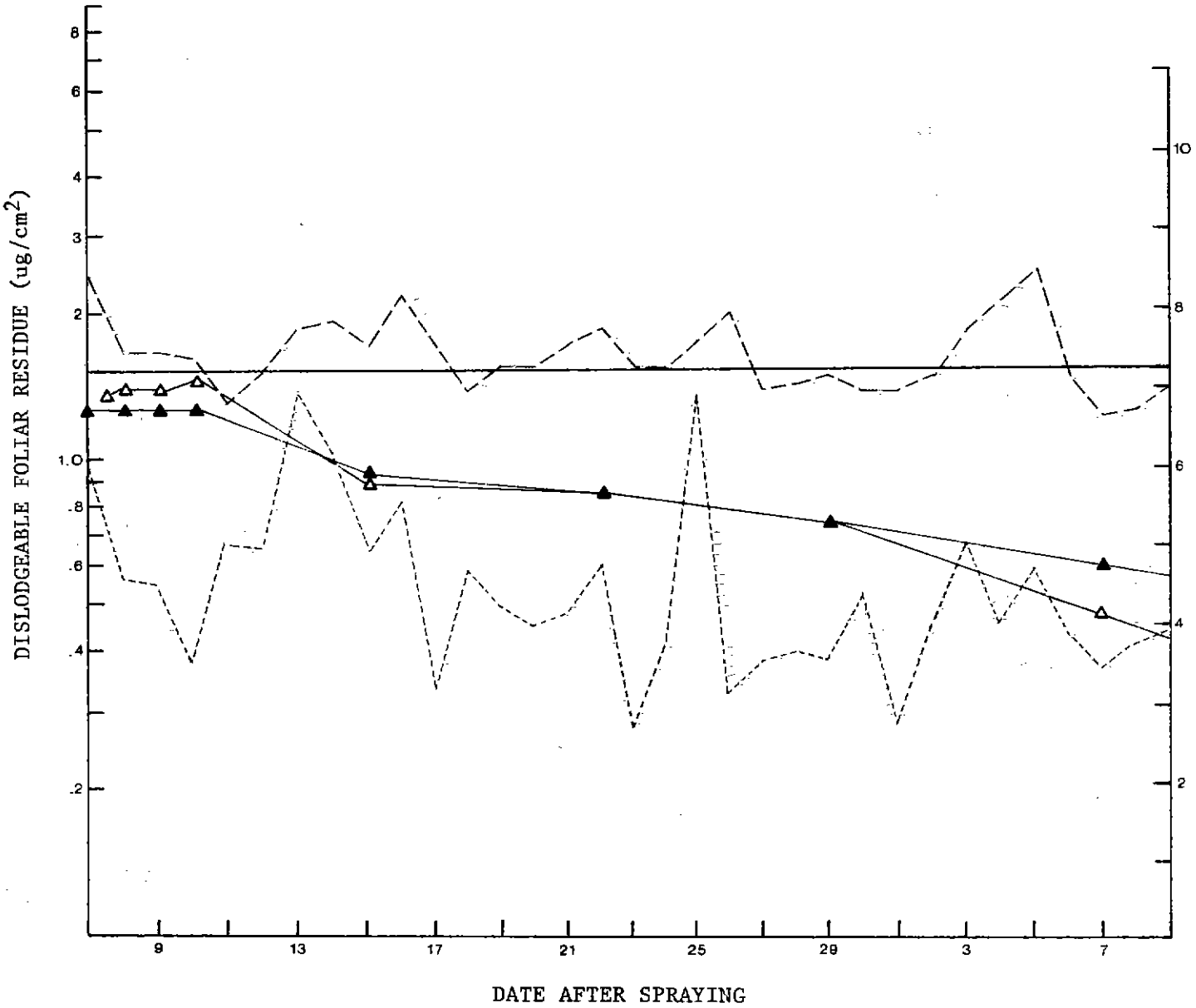

Figure 1 - Mean value degradation rates for dislodgeable Guthion plus Guthion oxon analogue (GOA) for the flood irrigated orchard (open symbols) and the sprinkler irrigated orchard (closed symbols) for Grower One, in ug/cm^2. The coarse dashed line is mean daily temperature in degrees Faherenheit x 10 using the right-hand scale. The fine dashed line represents total oxidants in parts per hundred million (PPHM), using the right-hand scale. The solid line at 1.6 ug/cm^2 on the left-hand scale is the calculated safe level for Guthion plus GOA.

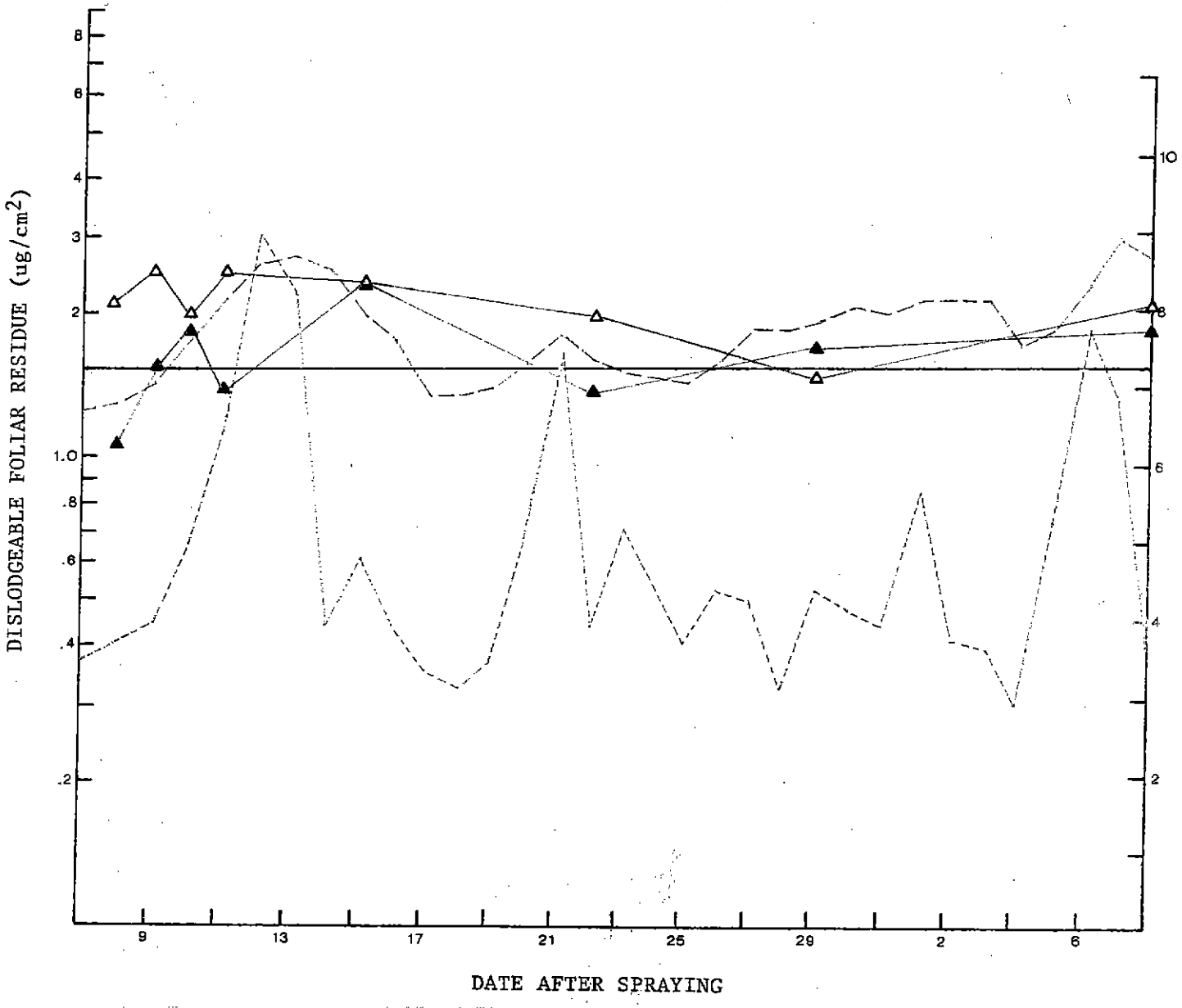

Figure 2 - Mean value degradation rates for dislodgeable Guthion plus Guthion oxon analogue (GOA) for the Monaco (open symbols) and McKnight (closed symbols) varieties of peaches of Grower Two, in ug/cm^2. Mean daily temperature (coarse dashed line) in degrees Farenheit x 10, and total oxidants (fine dashed line) in parts per hundred million (PPHM) use the right-hand scale. The solid line at 1.6 ug/cm^2 on the left-hand scale is the calculated safe level for Guthion plus GOA.

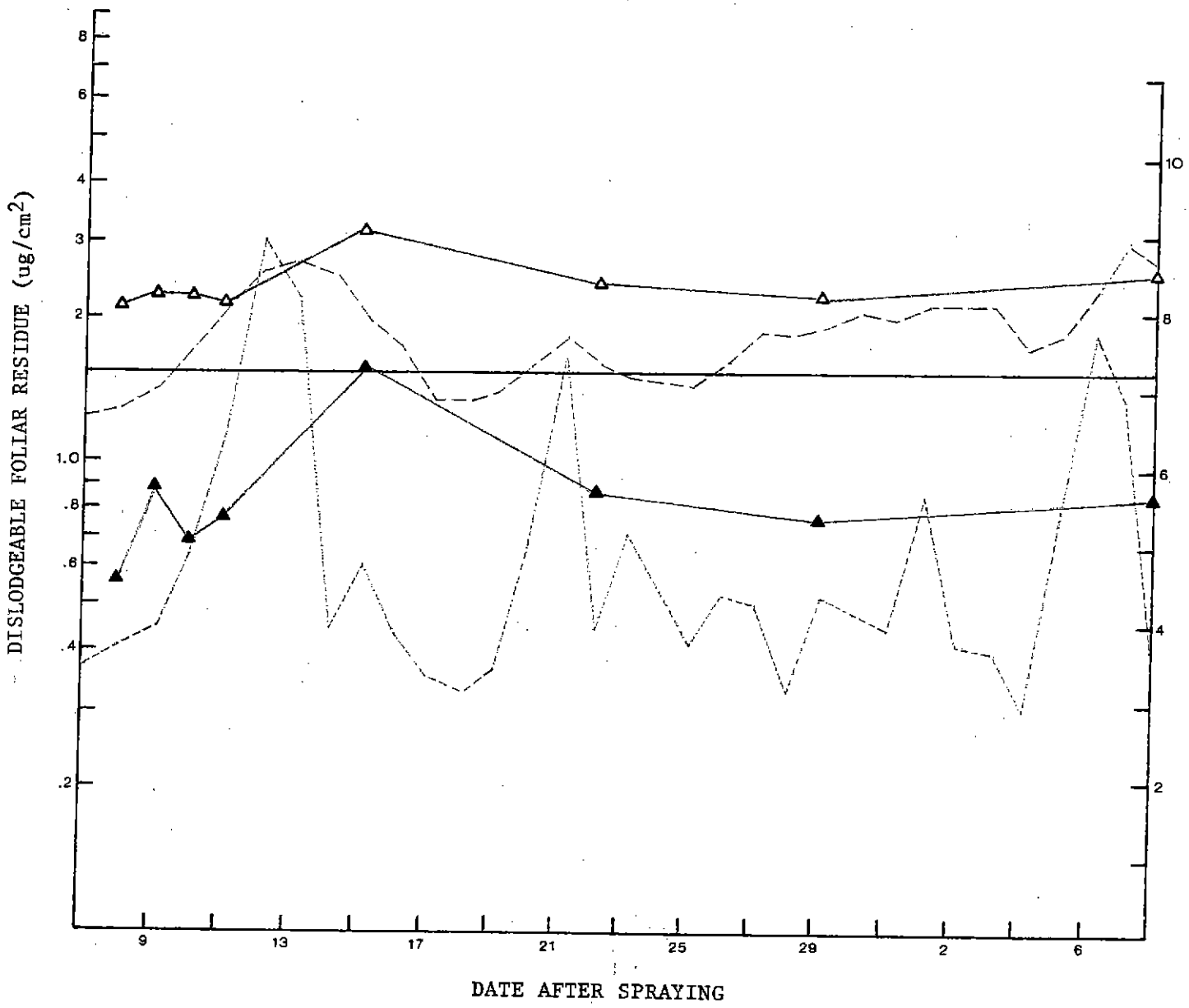

Figure 3 - Mean value degradation rates for dislodgeable Guthion plus Guthion oxon analogue (GOA) for the Caroline variety (open symbols) and the control (closed symbols) peaches of Grower Two, in ug/cm^2. Mean daily temperature (coarse dashed line) in degrees Farenheit x 10, and total oxidants (fine dashed line) in parts per hundred million (PPHM) use the right-hand scale. The solid line at 1.6 ug/cm^2 on the left-hand scale is the calculated safe level for Guthion plus GOA.

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